

Domain structures in ultrathin magnetic films

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Abstract

Domain structures appear in ultrathin magnetic films, if an easy-axis anisotropy, favouring the direction perpendicular to the film, overcomes the shape anisotropy. We will study the nature of the magnetic ground state and how the different parameters influence the domain structure. Both Ising and Heisenberg spins will be considered: in the latter case, a reorientation phase transition may take place. Domains are expected to appear just before such transition. Domain structures at finite temperature will be described in terms of a two dimensional array of domain walls, whose behaviour resembles a liquid crystal.

1 Introduction

A three dimensional ferromagnetic sample, below its Curie temperature, generally has no net magnetization because –as suggested by Weiss [1] and discovered by Bitter [2]– it is made up of contiguous regions where the magnetization is oriented along different directions. These regions (typical size $\approx 1 \div 10\mu\text{m}$) are called magnetic domains, and are separated by domain walls (typical size $\approx 10^2 \div 10^3\text{\AA}$), where the magnetization rotates continuously from one direction to another.

The magnetization inside each domain is due to the ferromagnetic coupling (\mathcal{H}_{ex}) between neighboring spins, which aligns them parallelly. The existence of domains is due to the dipolar energy (\mathcal{H}_{dip}): the true “magnetic” (and long-range) interaction between spins. This energy is also called “magnetostatic energy” or “shape anisotropy”: in fact, its minimization strongly depends on the shape of the sample. Finally, domains are not magnetized in random directions, but they prefer certain directions, depending on the crystal-axes, whose existence is due to the so-called magnetocrystalline anisotropies (\mathcal{H}_{an}). The actual magnetic structure of a system depends on the minimization of the total energy $\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{dip}} + \mathcal{H}_{\text{an}}$.

In this paper, we will be interested in ultrathin magnetic films: by “ultrathin” we mean a thickness smaller than, or of order of $N = 10$ atomic monolayers. Such small thicknesses can be currently obtained through growth by Molecular Beam Epitaxy [3]. Our aim is to give a microscopic description of domain structures in such two-dimensional systems, both at zero temperature (Section 2) and at finite temperature (Section 5). Section 3 is devoted to the dependence of the domain size L on the film thickness N . A comparison with Kittel’s theory –valid for much thicker films ($N > L$)– will be done.

Ultrathin films can undergo a reorientation transition, with the magnetization passing from being perpendicular to the film to being parallel to it. Since the condition for the occurrence of this transition coincides with the condition for having domains not unphysically large, the interplay between reorientation transition and magnetic domains will be treated in Section 4.

2 Domain structures at zero temperature

The aim of the present section will be the study of the classical ground state of a ferromagnetic ultrathin film, in presence of dipolar interactions and of a uniaxial anisotropy. Here and in the following, both the spin modulus $|\vec{S}|$ and the lattice constant a of the cubic crystal will be put equal to one.

The hamiltonian of a single monolayer will generally be assumed to have the form

$$\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{an}} + \mathcal{H}_{\text{dip}} \quad (1)$$

where the first term

$$\mathcal{H}_{\text{ex}} = \frac{J}{2} \int d\vec{x} (\nabla \vec{S})^2 \quad (2)$$

is the Heisenberg interaction, in the continuum approximation. The second term

$$\mathcal{H}_{\text{an}} = -\frac{K}{2} \int d\vec{x} S_z^2(\vec{x}) \quad (3)$$

is a single-ion anisotropy caused by the crystal field; it appears because, in a thin film, the \hat{z} direction (perpendicular to the film) is not equivalent to the in-plane directions. The constant K may be negative (easy-plane effect), but in this case no domain structure appears. Therefore, only the opposite case ($K > 0$) will be considered. Finally, the third term

$$\mathcal{H}_{\text{dip}} = \mathcal{H}_{\text{dip}}^{(1)} + \mathcal{H}_{\text{dip}}^{(2)} \quad (4)$$

is the dipolar interaction, which can conveniently be split into a rotationally invariant part

$$\mathcal{H}_{\text{dip}}^{(1)} = \frac{\Omega}{8\pi} \int \frac{d\vec{x}d\vec{x}'}{|\vec{x} - \vec{x}'|^3} \vec{S}(\vec{x}) \cdot \vec{S}(\vec{x}') \quad (5)$$

and an anisotropic part

$$\mathcal{H}_{\text{dip}}^{(2)} = -\frac{3\Omega}{8\pi} \int \frac{d\vec{x}d\vec{x}'}{|\vec{x} - \vec{x}'|^3} (\vec{S}(\vec{x}) \cdot \vec{\nu})(\vec{S}(\vec{x}') \cdot \vec{\nu}) \quad (6)$$

which couples the spin space to the “real” space. In addition to the constant Ω , we have introduced the notation: $\vec{\nu} \equiv (\vec{x} - \vec{x}')/|\vec{x} - \vec{x}'|$.

Since a spin doesn’t interact with itself, the integration must exclude a region around $x = x'$. This problem doesn’t exist if a discrete calculation replaces the integration (see below), or in the calculations relevant for domain structures, where x and x' are always far away (because belonging to different domains).

The term $\mathcal{H}_{\text{dip}}^{(1)}$ represents a long-range, antiferromagnetic coupling, while $\mathcal{H}_{\text{dip}}^{(2)}$ introduces an easy-plane anisotropy. In fact, this term vanishes for a perpendicular collinear state

(hereafter called \perp state), because $\vec{S} \cdot \vec{\nu} \equiv 0$, whilst for a parallel collinear state (hereafter called \parallel state), it has the negative value:

$$-\frac{3\Omega}{8\pi} \sum_{\vec{x}} \frac{(\vec{S} \cdot \vec{\nu})^2}{|\vec{x}|^3} \quad (7)$$

per spin. So, the \parallel configuration is energetically favoured with respect to the \perp one; furthermore, since only the in-plane component \vec{S}_{\parallel} of the spin contributes to it, for a generic collinear configuration forming an angle θ with the \hat{z} axis, $(\vec{S} \cdot \vec{\nu})^2$ rewrites as $(\vec{S}_{\parallel} \cdot \vec{\nu})^2$ and therefore expression (7) simply needs to be multiplied by $\sin^2 \theta$. So, for a collinear configuration, \mathcal{H}_{dip} is found to reduce –up to a constant– to the anisotropic expression:

$$\mathcal{H}_{\text{dip}}^{\text{an}} = -\frac{\Omega}{2} \sin^2 \theta \int d\vec{x} = \frac{\Omega}{2} \int d\vec{x} S_z^2 \quad (8)$$

Sometimes, instead of Ω we will use the quantity $g \equiv \Omega/16\pi$, which appears in the discrete version of dipolar interaction:

$$\mathcal{H}_{\text{dip}} = g \sum_{i>j} \frac{1}{r_{ij}^3} [\vec{S}_i \cdot \vec{S}_j - 3(\vec{S}_i \cdot \nu)(\vec{S}_j \cdot \nu)] .$$

Because of the competition between $\mathcal{H}_{\text{dip}}^{(1)}$ (a weak long-range antiferromagnetic (AFM) coupling) and \mathcal{H}_{ex} (a strong short-range ferromagnetic (FM) coupling), a domain structure arises, for *any* value of the dipolar coupling; furthermore, the competition between $\mathcal{H}_{\text{dip}}^{(2)}$ (an effective easy-plane anisotropy) and \mathcal{H}_{an} (an easy-axis anisotropy) can produce a re-orientation phase transition (RPT) of the magnetization, from the \perp state to the \parallel state. To prove the former statement, it is sufficient to show that the \perp state is destabilized by the introduction of an Ising domain wall. So, if we invert in the negative \hat{z} direction all the spins in the half-plane $x > 0$, we have to compare the cost of the domain wall ($2J$ per unit length) with the energy gain, due to dipolar interaction. Such gain is obtained by summing up $(-2g/|\vec{x} - \vec{x}'|^3)$ on all the spins (\vec{x}') of the half-plane $x > 0$ and the spins (\vec{x}) of the half-plane $x < 0$, with $y = \text{constant}$ (because we need the gain per unit length). The first sum on \vec{x}' corresponds to two spatial integrations, giving a quantity of order $-2g/|x|$. Therefore, the third sum diverges logarithmically at large distances¹ whatever the value of g , thus proving that the dipolar gain always prevails on the exchange loss.

The previous rough calculation also allows to have a semi-quantitative expression for the domain size of a stripe domain structure. In fact, the third integration on x will be limited up to the domain size L , by giving an energy gain of order $-g \ln L$ which must be approximately equal to the domain wall energy (per unit length): $g \ln L \approx E_{dw}$. So:

$$L \approx \exp(E_{dw}/g) . \quad (9)$$

For an Ising system, $E_{dw} = 2J$, so that (at zero temperature: $T = 0$) the domain size is exponentially large, unless J and g are of the same order. In Section 5, we will see how the previous result is modified, if thermal effects are taken into account.

For an Heisenberg system, the domain wall energy has the form [4]: $E_{dw} = 2\sqrt{\lambda J}$, where λ is the *total* effective easy-axis anisotropy (see Eq. (8)): $\lambda = K - \Omega$. Therefore, domains of “small” extension can be obtained even if $J \gg K, \Omega$, provided that $K \simeq \Omega$.

¹At small distances, the lattice constant is the natural cut-off.

Our calculations will concern a square lattice. A triangular lattice gives rise to frustration, in presence of an antiferromagnetic exchange coupling. Indeed, dipolar coupling is an antiferromagnetic interaction, in the Ising limit; anyway, it is a long-range one: so, the induced frustration exists even in a square lattice. We expect that the picture discussed in the rest of this Section is not qualitatively modified for a triangular lattice, if $L \gg 1$.

2.1 Domain walls

The size of a domain wall is given by the balance between the effective easy-axis anisotropy, and the exchange energy. If the long-range character of \mathcal{H}_{dip} is neglected and only its easy-plane effect (8) is retained, it is possible to determine analytically the magnetization profile inside the wall, and the domain wall extension.

If the wall is parallel to the y axis (as it will be always supposed in the following), in the continuum approximation the domain wall energy (per unit length) is

$$\mathcal{H}_{dw} = \frac{1}{2} \int dx \left[J(\nabla \vec{S})^2 - \lambda S_z^2 \right], \quad (10)$$

which must be minimized with respect to $\vec{S}(x)$.

By using polar coordinates ($\vec{S} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$) we obtain

$$\mathcal{H}_{dw} = \int \frac{dx}{2} \left\{ J[(\partial_x \theta)^2 + (\partial_x \phi)^2 \sin^2 \theta] - \lambda \cos^2 \theta \right\}. \quad (11)$$

The ϕ -dependent term is minimized when ϕ is constant, e.g. $\phi = \pi/2$ (Bloch wall) or $\phi = 0$ (Néel wall). On the other side, the minimization with respect to θ gives $2\partial_{xx}^2 \theta = (\lambda/J) \sin(2\theta)$, whose solution gives $\theta(x) = 2 \arctan(e^{x/w})$, where $w = \sqrt{J/\lambda}$. The corresponding expression of E_{dw} is obtained from Eq. (10): $E_{dw} = J/w + w\lambda = 2\sqrt{\lambda J}$.

The degeneracy among different types of wall is removed if dipolar interaction is correctly taken into account. It is immediately seen that the relevant interaction is the self-energy of the domain wall: in fact, let us consider the dipolar interaction between a spin \vec{S}_1 belonging to the wall, and a spin \vec{S}_2 belonging to a neighbouring domain. Since $\vec{S}_2 \cdot \vec{\nu} = 0$ ($\vec{\nu} = \vec{r}/r$, \vec{r} being the vector joining the two spins) only the isotropic interaction $g(\vec{S}_1 \cdot \vec{S}_2)/r^3$ is not vanishing. Anyway, the quantity $\vec{S}_1 \cdot \vec{S}_2 = \cos \theta_1$ does not discriminate among different types of wall, because it only depends on the axial angle θ_1 of the spin \vec{S}_1 , whilst walls differ in the polar angle ϕ_1 .

The dipolar contribution to the domain wall energy therefore writes:

$$E_{dw}^{dip} = \frac{g}{2} \sum_{x=0}^w \sum_{x'=0}^w \sum_{y=-\infty}^{\infty} \left\{ \frac{\vec{S}(x) \cdot \vec{S}(x')}{[(x-x')^2 + y^2]^{3/2}} - 3 \frac{(\vec{S}(x) \cdot \vec{r})(\vec{S}(x') \cdot \vec{r})}{[(x-x')^2 + y^2]^{5/2}} \right\}, \quad (12)$$

where we have used the translational invariance in the \hat{y} direction, and the following notations are understood: $\vec{r} = (x-x', y)$ and $\vec{S}(x) = (\sin \theta(x) \cos \phi, \sin \theta(x) \sin \phi, \cos \theta(x))$. The function $\theta(x)$ is any increasing monotonic function, such that $\theta(0) = 0$ and $\theta(w) = \pi$. Our aim is to minimize E_{dw}^{dip} with respect to the polar angle ϕ .

It is straightforward to obtain the following relations:

$$\begin{aligned} \vec{S}(x) \cdot \vec{S}(x') &= \cos(\theta(x) - \theta(x')) \\ (\vec{S}(x) \cdot \vec{r})(\vec{S}(x') \cdot \vec{r}) &= \sin \theta(x) \sin \theta(x') [(x-x')^2 \cos^2 \phi + y^2 \sin^2 \phi + 2y(x-x') \sin \phi \cos \phi] \end{aligned}$$

By retaining only the ϕ -dependent terms, we have:

$$E_{dw}^{dip} = -\frac{3g}{2} \sum_{x=0}^w \sum_{x'=0}^w \sum_{y=-\infty}^{\infty} \frac{\sin \theta(x) \sin \theta(x')}{[(x-x')^2 + y^2]^{5/2}} \times \left\{ \sin^2 \phi [y^2 - (x-x')^2] + y(x-x') \sin 2\phi \right\}.$$

The term proportional to $\sin 2\phi$ gives a zero contribution, because it is an odd function of y . So, we are left with the expression:

$$\begin{aligned} E_{dw}^{dip}(\phi) &= -\frac{3g}{2} \sin^2 \phi \sum_{x=0}^w \sum_{x'=0}^w \sum_{y=-\infty}^{\infty} \sin \theta(x) \sin \theta(x') \frac{y^2 - (x-x')^2}{[(x-x')^2 + y^2]^{5/2}} \\ &\equiv -\frac{3g}{2} \sin^2 \phi \cdot \mathcal{I}(w) \end{aligned}$$

Generally speaking, the shape anisotropy due to dipolar interaction implies that two spins $\vec{S}(\vec{x})$, $\vec{S}(\vec{x}')$ will prefer to align parallelly to $(\vec{x} - \vec{x}')$. Therefore the sum $\mathcal{I}(w)$ simply reflects the shape anisotropy of a stripe: $\overline{y^2} > \overline{(x-x')^2}$, where the bar means some average on the stripe. In other words, the dipolar energy of the domain wall is minimized if the spins point on the average in the (elongate) \hat{y} direction. The positive factor $\sin \theta(x) \sin \theta(x')$ does not play a major role: it is present because the in-plane component of the spin varies inside the wall. In conclusion, $\mathcal{I}(w)$ is positive and E_{dw}^{dip} is minimal for $\phi = \pi/2$, i.e. for a Bloch-type wall.

2.2 The Ising model

Once we have shown that the ferromagnetic configuration is destabilized by dipolar interactions, whatever the value of g , in this section we will study more closely the Ising model, in the entire range of values for the ratio g/J . Let us start with the physically relevant case $g/J \ll 1$, which corresponds to very large domains.

By extending a little more the naïve calculation made in the introduction, it is possible to determine the *leading* term in the expression of the dipolar energy of a stripe configuration. In order to use the following results also for the Heisenberg model, and for a larger number of atomic planes, we will suppose the two neighbouring stripes of size L to be separated by a domain wall of size $w \ll L$ and to lie on different planes at distance c . So, the relevant integral is the following:

$$I(c) = - \int_0^{L-w} dx' \int_0^L dx \int_{-\infty}^{+\infty} dy \frac{1}{[(x+x')^2 + y^2 + c^2]^{3/2}}, \quad (13)$$

whose general solution is:

$$\begin{aligned} I(c) &= \left(\frac{4L-2w}{c} \right) \arctan \left(\frac{2L-w}{c} \right) - \ln \left[1 + \left(\frac{2L-w}{c} \right)^2 \right] - \left(\frac{4L}{c} \right) \arctan \left(\frac{L}{c} \right) \\ &\quad + 2 \ln \left(1 + \frac{L^2}{c^2} \right) + \frac{2w}{c} \arctan \left(\frac{w}{c} \right) - \ln \left(1 + \frac{w^2}{c^2} \right). \end{aligned} \quad (14)$$

In the case $c = 0$ (corresponding to a single monolayer) or for $w \gg c$, we find $I(c) \approx 2 \ln(L/2w)$. In these cases, the dipolar energy gain with respect to the \perp state is, for a couple of planes at distance c :

$$E_{dip}^{2 \text{ planes}} = -2gI(c) = -4g \ln \left(\frac{L}{2w} \right). \quad (15)$$

For an Ising monolayer, $c = 0$ and $w = 1$, the total energy per spin of a stripe (S) configuration with respect to the \perp state is:

$$E^S(L) = \frac{2J}{L} - \frac{4g}{L} \ln(L/2) . \quad (16)$$

However, the factor 2 inside the logarithm (which modifies, in a certain sense, the value of J : $J \rightarrow J + 2g \ln 2$) is not meaningful, because it contributes to the non leading term of the dipolar energy. Conversely, it is noteworthy that the leading term (proportional to $g \ln L$) is the one obtained by considering the interaction (per unit length) of a stripe of size L with a neighboring stripe of infinite size! Analogously, the same leading term appears if the neighbouring stripe is indeed a square of size L . This result therefore provides –in the same approximation– the expression $E^C(L)$ for the energy of a square network (C=chess-board) of size L :

$$E^C(L) = \frac{4J}{L} - \frac{8g}{L} \ln(L) \equiv 2E^S(L) . \quad (17)$$

Therefore, not only the exchange cost of a square network is the double of a stripe structure with the same period (as the total length of domain walls is the double), but also the dipolar gain is the double. In very poor terms, the reason is that the infinite size of a stripe contributes only to the next-to-leading order term. Since in a square network each domain has four neighboring domains with opposite magnetization, whilst in a stripe configuration only two, we have the result $E^C(L) = 2E^S(L)$ (at the leading order in the dipolar energy!).

The previous results have been found by Czech and Villain [5] (see their Eq. (12)), who have drawn the erroneous conclusion that the chess-board configuration has a lower energy than the stripe configuration. Indeed, the next-to-leading order terms modify the previous picture, as explained by Kaplan and Gehring [6]. Through a more careful evaluation of the dipolar sums, they find that $E^S(L)$ and $E^C(L)$ are modified as follows:

$$\begin{aligned} E^S(L) &= \frac{2J - \alpha_S g}{L} - \frac{4g}{L} \ln L \\ E^C(L) &= \frac{4J - \alpha_C g}{L} - \frac{8g}{L} \ln L \end{aligned}$$

where $\alpha_S \simeq 4.2$ and $\alpha_C \simeq 0.3$. By minimization with respect to L , we find

$$\begin{aligned} L_S &= \exp \left[\frac{J}{2g} + 1 - \frac{\alpha_S}{4} \right] & E^S(L_S) &= -\frac{4g}{L_S} \\ L_C &= \exp \left[\frac{J}{2g} + 1 - \frac{\alpha_C}{8} \right] & E^C(L_C) &= -\frac{8g}{L_C} \end{aligned}$$

From the previous expressions, it is apparent why neglecting terms of order (g/L) in the energy leads to mistakes. Furthermore, $E^S < E^C$ if $L_C/L_S > 2$, that is to say $(\alpha_S/4 - \alpha_C/8) > \ln 2$, a relation which is satisfied.

Now, we will consider the opposite limit of a large ratio g/J : dipolar interaction prevails over the FM exchange. In the following, the notations C- L and S- L will mean, respectively a square network of side L and a stripe configuration of period L . Since dipolar interaction is a (long-range) AFM coupling, we are led to conclude that the AFM state (i.e. the C-1 state) is the lowest energy one: an explicit numerical calculation [7] shows that the C-1 state is indeed the ground state.

Such a state is maintained for a sufficiently weak J . So, a square network is preferred at (very) high g/J , but stripe configurations are preferred at low g/J . We can ask when the transition between square configurations and stripe ones takes place. According to numerical results [7], for $J \simeq 0.4g$, the C-1 state is replaced by the S-1 state, and for increasing J we simply have an augmentation of the period L of the stripe configuration. Since analytical theories are not applicable here, let us give some simple arguments to justify why C-1 is replaced by S-1, rather than C-2. Let us compare the energies of the two possible configurations:

$$\begin{aligned} E^C(2) &= E_{dip}^C(2) + 4J/2 \\ E^S(1) &= E_{dip}^S(1) + 2J . \end{aligned}$$

So, it is sufficient to compare their dipolar energies. Both the configurations have a vanishing nearest bond contribution to the dipolar energy (two spins up and two spins down), but in the S-1 state all the four nn-bonds are AFM, whilst in C-2 are half and half. For the 3rd nearest bonds, the picture is reversed, but the coupling is weaker, and up to the 6th nearest bonds the contributions are the same. This simple evaluation gives the correct result.

We conclude this section by summing up the main results. Dipolar interactions favour an AFM coupling between spins; if we consider chess-board (C- L) and stripes (S- L) configurations, we have that the gains of dipolar energy (with respect to the \perp state) $|E_{dip}^S(L)|$ and $|E_{dip}^C(L)|$ are decreasing functions of L and $|E_{dip}^C(L)| > |E_{dip}^S(L)|$. Nevertheless, when a FM exchange interaction is taken into account, the stripe configuration is energetically favoured, except for $J/g < 0.4$, when the ground state is the AFM one.

2.3 The Heisenberg model

In the previous section we have supposed that spins are “Ising spins”: \vec{S} is always parallel to the \hat{z} axis. Now spins are allowed to rotate, and the main difference is the energy of the domain wall, which is lowered with respect to the Ising case. This allows smallest domain sizes, even if –at $T = 0$ – they will be seen to remain of macroscopic extent, except in a very narrow region of the parameter space. A second important feature of the ferromagnetic Heisenberg model, is that its excitations are spin-waves, which allow a local linear stability analysis.

Some insight in the model is given by the study of the collinear configurations, the generic one forming an angle θ with the \hat{z} axis. Since the total effective uniaxial anisotropy is $\lambda = (K - \Omega)$, the \perp state ($\theta = 0$) will be the lowest energy one for $K > \Omega$. For $K < \Omega$ the magnetization will lie in the plane ($\theta = \pi/2$), and for $K = \Omega$ all the collinear states will be degenerate.

The previous section has been devoted to show (for an Ising model) how the long-range character of the dipolar interaction “destabilizes” the \perp state. This result is certainly not modified for an Heisenberg model, which simply allows for a much larger phase space. Anyway, there is no reason to think that an infinitesimal variation in the \perp state will lead to a lower energy: in other words, the \perp state may be locally stable and spin-waves do not destabilize it, i.e. it may be a metastable state. In fact, the dispersion curve writes:

$$\omega_{\perp}(q) = (K - \Omega) + Jq^2 = \lambda + Jq^2 . \quad (18)$$

Dipolar interaction simply modifies the gap, because of its “easy-plane” effect.

If now we turn to the \parallel state ($K < \Omega$), a first question is: Do dipolar interactions destabilize the \parallel state? The answer is no: this case, domains are not energetically favoured. In the \perp state, domains rise because of the competition between the FM \mathcal{H}_{ex} and the AFM $\mathcal{H}_{\text{dip}}^{(1)}$. Anyway, in the \parallel state, the anisotropic term $\mathcal{H}_{\text{dip}}^{(2)}$ (which vanishes almost everywhere in the \perp state) changes the character of \mathcal{H}_{dip} . Let us compare the in-plane FM and AFM configurations.

The spin in the origin is choosen along the \hat{y} axis: this way, all the other spins can be arranged in circular rings of increasing radius where spins are mutually parallel, and –in the FM state– always oriented along the \hat{y} direction, or –in the AFM state– alternately oriented along \hat{y} and $-\hat{y}$. An in-plane domain structure may arise only if \mathcal{H}_{dip} favours the AFM configuration with respect to the FM one. To check it, let us consider the dipolar interaction between the spin i in the origin and all the spins j on a given circular ring at distance r . The resulting sum is proportional to $\pm[1 - 3\langle(\vec{S} \cdot \vec{v}_{ij})^2\rangle_j]/r^3$, where the sign $+$ (resp. $-$) holds for the FM (resp. AFM) configuration. Since the average $\langle \dots \rangle_j$ on the spins j gives $(1/2)$, the negative anisotropic part prevails on the first one, and the energy is lower for the FM state! Consequently, domains are not expected if the magnetization lies in the film plane.

Anyway, the linear stability analysis of the \parallel state reveals important features. In fact, it was shown by Maleev [8] in the seventies that dipolar interactions are able to stabilize the \parallel state at finite temperature, because of their long-range character. This means that the dispersion curve is modified well beyond the introduction of some effective easy-plane anisotropy, which is unable to assure a finite magnetization. The correct expression for the dispersion curve is [8, 9, 10] ($\theta_{\vec{q}}$ is the in-plane angle between \vec{q} and the magnetization):

$$\omega_{\parallel}^2(q) = \left[(\Omega - K) - \frac{\Omega}{2}q + Jq^2 \right] \times \left[\frac{\Omega}{2}q \cos^2 \theta_{\vec{q}} + Jq^2 \right] \equiv \omega_{\text{out}} \times \omega_{\text{in}} . \quad (19)$$

We have assigned a different meaning to the two terms, because now the Goldstone mode ($\omega(q=0) = 0$) exists for ω_{in} , but not for ω_{out} . Anyway, an out of plane excitation with a *finite* wavevector may go to zero and destabilize the \parallel configuration. This happens for $\bar{q} = \Omega/4J$, when $1 > (K/\Omega) > 1 - (\Omega/16J)$. So, for a weak dipolar interaction such interval is extremely narrow.

We can summarize the results of the spin-wave stability analysis of the FM configurations, as follows. The \perp state is locally stable, and therefore it is metastable, because the insertion of a domain wall lowers the energy of the system. Domains are not expected in the in-plane state; nonetheless, the long-range character of \mathcal{H}_{dip} destabilizes the \parallel configuration, if Ω is just a bit larger than K . The main result of the next section will be to link quantitatively the results of the previous analysis, with energetic calculations on domain structures.

2.4 Yafet and Gyorgy's theory

To my knowledge, the only microscopic study of domain structures in a Heisenberg system, is due to Yafet and Gyorgy [11]. They study a stripe domain structure, characterized by two parameters: the domain size L and the domain wall size w . The wall is of Bloch-type, and the component S_z of the magnetization, along the domain wall, has a sinusoidal profile. As suggested by the same authors, we have added a third parameter which reveals essential to match the results of the spin-wave stability analysis: the maximal component s of the

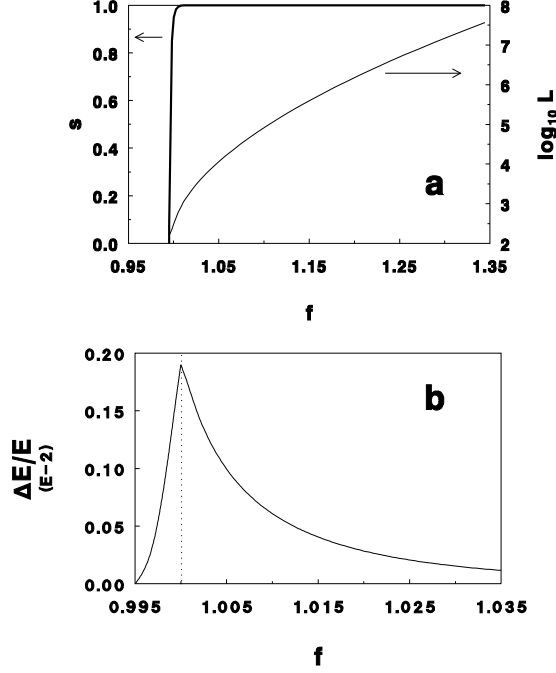


Figure 1: (a) The maximal value s of the component $|S_z|$, inside each domain, and the domain size L (in logarithmic scale). (b) The energy gain of the domain structure, relative to the collinear state (the \perp one for $f > 1$ and the \parallel one for $f < 1$). The smallest value of f shown in (b) is f_m , to which corresponds—in (a)—the vanishing of s . For both figures, $2L_d = 25$. Similar dependence for $L(f)$ and (b) had already been given in Ref. [11].

magnetization in the \hat{z} direction, inside each domain. $s = 0$ means the \parallel configuration, while $s = 1$ is a “standard” domain structure. The energy of a generic stripe domain structure is therefore a function of the three variational parameters (L, w, s) . Its minimization, and a comparison with the energy of a generic collinear state, will allow to determine the lowest energy configuration.

In Fig. 1 we plot (a) L and s , as a function of the ratio $f \equiv K/\Omega$, and (b) the energy gain of the domain structure. Two features are manifest: (i) It exists a minimal value $f_m < 1$, beyond which a domain structure is energetically favoured. As expected, the energy gain persists till $f = \infty$ and it is maximal for $f = 1$. (ii) When $f = f_m$, spins lie in the plane ($s = 0$), and $L = L_m = 4\pi L_d$, where $L_d = J/\Omega$ is the dipolar length. Upon increasing f , the maximal component of S_z immediately saturates to $s = 1$, and the domain size L grows exponentially: unless dipolar coupling and uniaxial anisotropy nearly compensates, domains are unphysically large. How close to one f must be, in order to observe domains, it depends on the dipolar length. It is not possible to give here the details of the calculation, which follow Ref. [11]. Anyway, the introduction of the parameter s modifies the values of f_m and L_m found by Yafet and Gyorgy, which now match the corresponding values, determined *via* the linear stability analysis of the \parallel configuration [12, 13]: $f_m = 1 - (\Omega/16J)$ and $L_m = \pi/\bar{q}$ (see below Eq. (19)).

In conclusion, the following picture emerges: If K is fairly smaller than Ω , the \parallel configuration is the ground state; when K is nearly equal to Ω ($K = \Omega - \Omega^2/16J$) a soft mode

appears, with a finite wavevector $\bar{q} = \pi/L_m$. In the new configuration, spins acquire a component S_z which is alternately positive and negative. Upon further increase of K , a domain structure clearly appears, with regions where $S_z = \pm 1$ are separated by domain walls of size w . These configurations are the true ground state, *but* for $K > \Omega$ the \perp state is locally stable. The domain size L increases exponentially and rapidly becomes larger than the sample size.

The main difference between Ising and Heisenberg models is the form, and therefore the cost, of a domain wall: in the former case, the wall size is one lattice constant, and its energy per unit length is simply $E_{dw}^{Ising} = 2J$. In the latter case, the wall can extend over many lattice constants and its cost is given by the balance between easy-axis anisotropy and exchange: $E_{dw}^{Heisenberg} = 2\sqrt{J(K - \Omega)}$. This expression correctly shows that the domain wall energy goes to zero, when K and Ω nearly compensate. Nevertheless, it has the drawback not to match E_{dw}^{Ising} when $K \rightarrow \infty$: this is because it derives from a continuum approximation, which fails to reproduce the discontinuity of an Ising domain wall. However, when the Ising limit is pertinent, domains are very large and probably unphysical. So, this limit is not really relevant (at least at $T = 0$).

An approximate analytical expression for L can be found by noticing that the dipolar interaction between domains can be evaluated –in the Heisenberg model– by “removing” the domain wall [14]: so, we have Ising domains left, at distance w . By using Eq. (14) and the expression given in Section 2.1 for the energy of a domain wall, we have the following formula for the energy per spin of a stripe domain configuration:

$$E_{dom} = \frac{\lambda w}{L} + \frac{J}{Lw} - \frac{4g}{L} \ln\left(\frac{L}{2w}\right), \quad (20)$$

which must be minimized not only with respect to L , but also w . The calculation gives:

$$\begin{aligned} w &= \frac{16\pi L_d}{2 \left[1 + \sqrt{1 + 4^3 \pi^2 L_d (f - 1)}\right]} \\ L &= 2w \exp(J/2gw). \end{aligned} \quad (21)$$

This expression for L is valid also for the Ising model, where $w = 1$. The form of w proves the existence of a minimum value f_m , for which the square root vanishes and $L \simeq L_d$. The value of f_m found *via* the approximate expressions given above is not the correct one: $f_m = 1 - 1/16L_d$, because Eq. (20) neglects the parameter s .

3 Domain size and film thickness

3.1 Kittel’s theory and modifications

The theories developed in the previous section apply to a single monolayer. Anyway, before extending them to a (thin) film of N planes, let us report Kittel’s old results [15], which are valid for a thick film, whose thickness is larger than the domain size ($N > L$), and a recent modification [16], introduced to take into account the possible surface character of the anisotropy K .

In Kittel’s theory, the total energy (F_{dom}) of the magnetic slab is the sum of three contributions: the magnetostatic one (F_{dip}), the domain-wall energy (F_{dw}), and the anisotropy term (F_{an}). He considered both the possibility of flux closure (Fig. 2a), and of an open

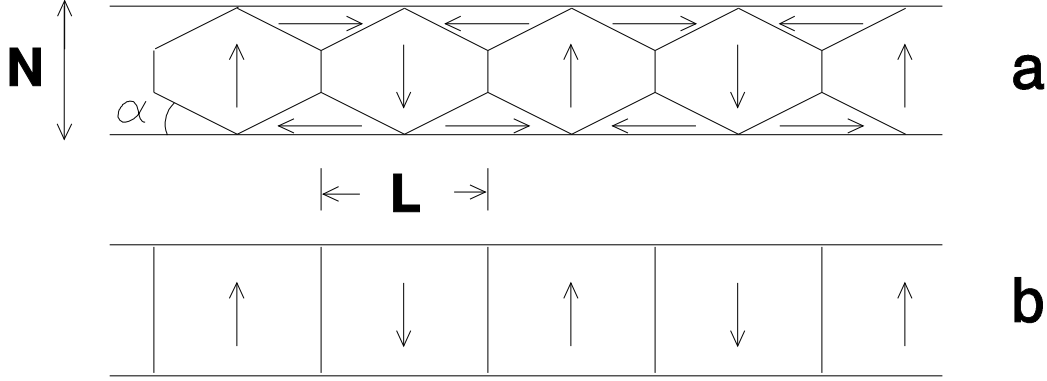


Figure 2: Domain structures in a thin film: flux closure (a) and open flux circuit (b).

flux circuit (Fig. 2b), because he was interested in determining the lower energy configuration. However, the dependence $L(N)$ is the same for the two cases. If closure domains are present, the contribution of the magnetostatic energy is neglected and the anisotropy term is proportional to the volume of the domains where the magnetization lies in the film plane. Finally, F_{dw} depends on the extension of the domain walls. If σ_{w1} and σ_{w2} are the wall energies per unit area, respectively of the 90° and 180° walls, the total energy $F_{dom}^{(a)}$ (per unit of surface) of the closure domain structure, writes²:

$$F_{dom}^{(a)} = F_{dw}^{(a)} + F_{an}^{(a)} \equiv \left[\frac{2\sigma_{w1}}{\cos \alpha} + \sigma_{w2} \left(\frac{N}{L} - \tan \alpha \right) \right] + K \frac{L}{2} \tan \alpha . \quad (22)$$

Kittel considered the limit $N \gg L$ and assumed $\sigma_{w1} = \sigma_{w2}$ and $\alpha = 45^\circ$.

The minimization of $F_{dom}^{(a)}$ gives $L = \sqrt{2\sigma_{w2}N/K \tan \alpha}$ and $F_{dom}^{(a)} = (2\sigma_{w1}/\cos \alpha) - \sigma_{w2} \tan \alpha + \sqrt{2K \tan \alpha \sigma_{w2}N}$. So, the domain size is proportional to the square root of the film thickness.

Such a result is found also in absence of closure domains: in such case $F_{dom}^{(b)} = F_{dw}^{(b)} + F_{dip}^{(b)}$, because $F_{an}^{(b)} = 0$. Nevertheless, the relevant functional dependences are unchanged: $F_{dw}^{(b)} = \sigma_{w2}N/L$ and $F_{dip}^{(b)} \approx M_s^2 L$ (M_s being the magnetization of saturation), i.e. it has the same L -dependence of $F_{an}^{(a)}$. Consequently, the result $L \sim \sqrt{N}$ still holds.

Recently, the above calculation has been modified [16] to take into account the possibility of a surface anisotropy, in an ultrathin film. In this limit, $N \ll L$, closure domains are supposed to make a small angle α with respect to the surface of the film, and the total anisotropy K is written as the sum of a volume (K_v) and a surface (K_s) part: $K = K_v + 2K_s/N$. If $K_s (\approx 1 \text{ erg/cm}^2)$ prevails over $K_v (\approx 10^5 \text{ erg/cm}^3)$, the result by Kittel is modified as follows³:

$$L = \sqrt{\frac{2\sigma_{w2}N}{K \tan \alpha}} \simeq \sqrt{\frac{\sigma_{w2}}{K_s \tan \alpha}} N . \quad (23)$$

So, for a surface anisotropy, Kittel's theory gives a *linear* increase of the domain size with the thickness. This result agreed with some experimental findings [17] on thin epitaxial Co/Au(111) films. Nevertheless, the calculation in Ref. [16] does not seem to be consistent.

²The superscripts (a) and (b) refer, respectively to Fig. 2a and Fig. 2b.

³In Ref. [16], the trigonometrical factor $\tan \alpha$ is missing.

In fact, it is noteworthy that below a critical thickness, Kittel's theory predicts a single-domain in-plane state. This result is obtained by comparing the energy $F_{\parallel} = 2K_s + K_v N$ of a single-domain in-plane state with the energy of the closure domain state:

$$F_{dom}^{(a)} = \frac{2\sigma_{w1}}{\cos \alpha} + 2\sqrt{K_s \sigma_{w2} \tan \alpha} - \sigma_{w2} \tan \alpha . \quad (24)$$

If we write $\sigma_{w1} \simeq \sigma_{w2} \simeq \sigma_w$ and we neglect the last two terms with respect to the first one (because $\tan \alpha \ll 1$), we find that the in-plane magnetized state is energetically favoured if⁴ $N < 2(\sigma_w - K_s)/K_v$: a condition which is generally fulfilled in ultrathin films.

A more rigorous consideration is the following. Expression (22) for the energy $F_{dom}^{(a)}$ of a closure domain structure should be minimized not only with respect to L , but also to the angle α . If $\partial_{\alpha} F_{dom}^{(a)}$ is calculated, we obtain

$$\partial_{\alpha} F_{dom}^{(a)} = \frac{1}{\cos^2 \alpha} [(KL/2 - \sigma_{w2}) - 2\sigma_{w1} \sin \alpha] = 0 . \quad (25)$$

However, it is easily seen that its solution: $\sin \alpha = (KL/2 - \sigma_{w2})/2\sigma_{w1}$ does not correspond to a local minimum: the Jacobi matrix of the second derivatives has a positive and a negative eigenvalue, because $F_{dom}^{(a)}$ has a minimum with respect to L , but a maximum by varying α ! Indeed, this problem exists also for the original Kittel's theory, where α is arbitrarily put equal to 45° . A correct approach should probably add an α -dependent magnetostatic contribution $F_{dip}^{(a)}$.

3.2 Ultrathin film limit: microscopic approach

In the following, we will be concerned with the extension of the Ising and Heisenberg models to the case $N > 1$. Whilst the extension to $N > 1$ of the domain wall energy is straightforward for both models, the contribution of the dipolar interaction energy between different planes is not trivial, and some further approximation is necessary, in order to obtain tractable formulas.

Let us start with the Heisenberg model, which is even simpler than the Ising one. The domain wall size w varies in the interval $\sqrt{J/K} < w < J/g$. So, the hypothesis $w \gg N$ is reasonable, and in this limit the integral $I(c)$ in Eq. (14) does not depend on c . Such integral appears N times for the value $c = 0$ and $N(N-1)/2$ times for $c \neq 0$, that is to say a total of $N(N+1)/2$ times. Therefore, the expression (20) for the energy of the domain structure is modified as follows:

$$\frac{E_{dom}(N)}{N} \approx \frac{\lambda w}{L} + \frac{J\gamma(N)}{Lw} - \frac{2(N+1)g}{L} \ln \left(\frac{L}{2w} \right) . \quad (26)$$

The factor $\gamma(N)$, whose importance will be elucidated very shortly, has been introduced to take into account a possible "long-range" character of the exchange interaction. If the range r_{ex} of \mathcal{H}_{ex} is much smaller than N , then $\gamma(N) = 1$, but $\gamma(N) = \gamma^*(N) \equiv (N+1)/2$ if $r_{ex} > N$. Finally, the anisotropy term \mathcal{H}_{an} has been assumed to be of volume type.

The N -dependence of E_{dom} can be reduced to the following transformations: $J \rightarrow \gamma(N)J$ and $g \rightarrow \gamma^*(N)g$. The domain wall size w has no trivial transformation, but in the limit $L \gg w$, $w \rightarrow \sqrt{\gamma(N)}w$ (see Eq. 21). So:

$$L(N) \approx \sqrt{\gamma(N)}w_1 e^{\frac{\sqrt{\gamma(N)}}{\gamma^*(N)} \frac{J}{2gw_1}} , \quad (27)$$

⁴All the energies must be expressed in units where the lattice constant $a = 1$.

where w_1 refers to the monolayer ($N = 1$). Since $\sqrt{\gamma(N)}/\gamma^*(N)$ is always a decreasing function of N , a strong shrinkage of the domains is expected, even if \mathcal{H}_{ex} is “long-range”.

Now, let us consider the Ising model. When we deal with the dipolar term, it is no more possible to suppose $w \gg N$, because now $w = 1$. The integral (14) writes:

$$I(c) \simeq 2 + (2/c) \arctan(1/c) + 2 \ln(L/2\sqrt{1+c^2}) \approx 2 \ln(L/2\sqrt{1+c^2}) , \quad (28)$$

if $L \gg c$. The total dipolar energy gain E_{dip} is obtained by summing up $I(c)$ over c , for all the couples of planes. Kaplan and Gehring [6] have used a continuum theory by resorting to the magnetic potential: this corresponds to the limit $N \gg 1$. In this case $I(c) \approx 2 \ln(L/2c)$ and

$$\frac{E_{\text{dom}}}{N} \approx \frac{2J\gamma(N)}{L} - \frac{4Ng}{L} \ln\left(\frac{L}{N}\right) . \quad (29)$$

The factor N inside the logarithm does not modify the leading behaviour, but in opposition to the Heisenberg model, it becomes relevant if $\gamma(N) = \gamma^*(N)$. In fact, the minimization of (29) gives

$$L(N) \approx N e^{\frac{\gamma(N)}{N} \frac{J}{2g}} . \quad (30)$$

Now, if $\gamma(N) = \gamma^*(N)$, the exponential dependence on N disappears⁵ and the polynomial dependence becomes relevant: therefore, a linear increase of the domain size is predicted (in the Ising model!) if $r_{\text{ex}} > N$.

We can summarize the main results in the following way. In both models (Ising and Heisenberg models), L has an exponential dependence on $(J/2gw) \approx (E_{\text{dw}}/g)$. When the thickness is increased, in very rough terms $g \rightarrow \gamma^*(N)g$, but E_{dw} has a different expression in the two models: $E_{\text{dw}} \approx J \rightarrow \gamma(N)J$ in the Ising one, and $E_{\text{dw}} \approx \sqrt{J\lambda} \rightarrow \sqrt{\gamma(N)}E_{\text{dw}}$ in the Heisenberg one. So, in the limit $\gamma = \gamma^*$, the exponential dependence on N disappears in the Ising model, but it remains in the other one. This dependence implies that L decreases with N in the Heisenberg model, whilst in the Ising model, if the exponential doesn't depend on N , we have the pre-exponential dependence left: a dependence which makes L increase with N and which becomes a linear dependence in the continuum approximation.

We can ask how the result for the Heisenberg model is modified, if the anisotropy is of surface-type: since the domain wall energy decreases with N , narrower domains are expected! This result contradicts the findings on the closure domain structures [16], based on the modified Kittel's theory. The reason is the following: in this theory, the anisotropy contributes to the total energy through the closure domains, where the magnetization lies in the film-plane, whilst the domain-wall energy is tacitly supposed not to depend on it.

Instead, in our theory no closure domain is present –as expected, if the film is very thin– but the domain wall energy is proportional to the square root of the anisotropy.

The conclusion of the present section is that –as a general rule– in a thin film domains are expected to get narrower upon increasing the thickness. This behavior has been found experimentally by Speckmann et al. [18] in ultrathin Co/Au(111) films (above $N = 4$), by Bochi et al. [19] in Ni/Cu/Si(001) films, and by Gehanno and Samson [20] in ordered FePd thin films. So, a contradiction seems to exist between Ref. [17] and Ref. [18], both concerning Co/Au films. This contradiction might be solved guessing that the linear increase observed in Ref. [17] accords with the weak increase of Ref. [18], observed below $N = 4$. This behavior would correspond to the regime $N < r_{\text{ex}}$, which gives –for an Ising model!– an increasing

⁵In the limit $N \gg 1$, the distinction between N and $(N + 1)$ is not relevant.

$L(N)$. Above four monolayers, we entry in the regime $N > r_{ex}$ (a narrow regime which is missed in Ref. [17]), which is characterized by a strong decreasing of $L(N)$.

Anyway, the previous picture is not totally satisfactory, because it assumes the validity of the Ising model, or –in other words– of domain walls of atomic thickness, whilst walls extend over hundreds of lattice constants. Furthermore, all the previous “magnetic” models assume perfectly flat and periodic arrays of spins, even if defects are known to play a major role in determing domain structures. For example, Allenspach and Stampanoni [17] report an increase of the average lattice constant, upon increasing N . This means that dislocations exist in the film, and also that the exchange coupling constant is expected to decrease. Since J strongly determines the actual value of L , we can even suppose that the observed increase of $L(N)$ is indeed due to the lowering of J , and therefore of the domain wall energy.

4 Domain structures and reorientation transition

In the part of the previous section which was dedicated to the Heisenberg model, we shew the possible different ground states of an ultrathin ferromagnetic film, subject to an easy-axis anisotropy and to the dipolar interaction. In Fig. 3, we show the phase diagram at $T = 0$, in the space of the parameters $f = K/\Omega$, $L_d = J/\Omega$. Upon decreasing the ratio K/Ω , the system passes from a collinear \perp state, to a collinear \parallel state, across a domain structure.

As we have pointed out, the \perp state is never the ground state, in the thermodynamic limit. Anyway, from an experimental point of view, the question is different. First of all, domains may be even larger than the sample size, so that –in principle– the \perp state may be the true ground state of a finite system. Furthermore, our estimates of the domain size were valid at $T = 0$, for a perfect sample. Whatever the domain size is, the \perp state –which is metastable– is attained by the application of a magnetic field pulse: a typical procedure, before a magnetization measurement. Therefore, an important quantity is the relaxation time of the collinear state: if it is (much) longer than the measurement time, the \perp state may be considered as the “ground state”, whereas it will decay in a domain structure, in the opposite limit.

Experimentally, the phase diagram in the parameter space may be “visited” by varying the thickness and/or the temperature. In fact, the easy-plane effect of the dipolar interaction increases with N , whilst the easy-axis anisotropy generally keeps constant, because of its surface character. This means that the “effective” ratio K/g decreases, and indeed a transition from the \perp state to the \parallel one is observed, when the thickness is augmented. This transition is called reorientation phase transition (RPT), and if a microscopic probe is used to measure the magnetization (for example, the spin polarized scanning electron microscopy), magnetic domains are observed in the “transition” region between the two collinear states [21].

The same phenomenon of reorientation may be observed upon increasing the temperature, before the usual ferro-paramagnetic transition takes place. In this case, the theoretical study of the transition requires to take into account thermal fluctuations, which weaken the different coupling parameters (J, K, Ω). The RPT is the result of a strongest decrease of the uniaxial anisotropy, with respect to the dipolar coupling: so, $K(T)$ may be smaller than $\Omega(T)$ –and therefore the \parallel state the preferred one– even if $K(T = 0) > \Omega(T = 0)$. The T -dependence may be obtained through a perturbation theory [22, 23], or through a renormalization group analysis [24]. We will not enter into details here, nor we will give the

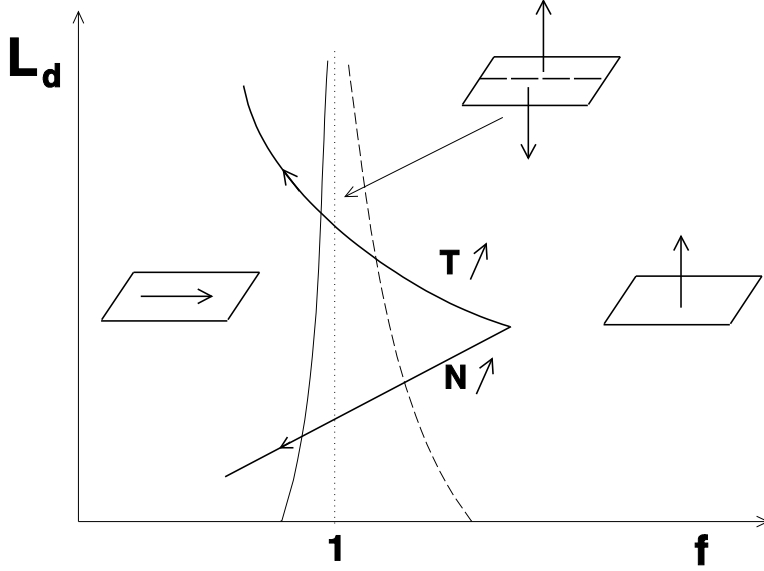


Figure 3: “Phase diagram” at $T = 0$, in the plane (f, L_d) . No units are shown on the f and L_d axis, because the diagram has only a qualitative meaning. The dotted line $f = 1$ corresponds to the separation between the \perp and \parallel FM configurations. By taking into account domain structures, domains rise in all the region on the right of the full thin line, but they are observable only on the left of the dashed line. So, starting from a thin film which is perpendicularly magnetized, we may observe a reorientation phase transition upon increasing the temperature, or upon increasing the thickness. In a region around $f = 1$, domains of observable size appear.

explicit dependences on T ; however, we want to discuss an important feature of the RPT: the order of the transition.

Firstly, let us address the question in the parameter space. If we consider the true ground state of the system, the answer is clear: the transition is continuous, because the domain structure (see Fig. 1a), in proximity to $f = 1$, evolves towards the \parallel state, passing through a shrinking of the domain size and the *continuous* change of s (the maximal value of $|S_z|$ inside each domain) from $s = 1$ to $s = 0$. This picture clearly shows the importance of introducing such variational parameter in the study of domain structures.

If domain structures are neglected, as in almost the whole body of theoretical papers on the RPT, the answer is more subtle, and –above all– it depends on details of the system. Limiting the investigation to collinear configurations corresponds, in the \perp phase, to study a metastable state⁶. Let us start with the simplest possibility: a single monolayer, subject to the Hamiltonian $\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{an}} + \mathcal{H}_{\text{dip}}$. We have shown that \mathcal{H}_{dip} has an effective easy-plane effect ($\mathcal{H}_{\text{dip}}^{\text{an}}$): as a consequence of this, the energy (per spin) of a collinear configuration writes:

$$E(\theta) = -(K - \Omega) \cos^2 \theta, \quad (31)$$

where θ is the angle between the magnetization and the \hat{z} axis. It is manifest that $E(\theta)$ allows only two possible minima, according to the sign of $(K - \Omega)$: $\theta = 0$ and $\theta = \pi/2$. So, in the parameter space, the transition is obviously discontinuous, because no canted configuration

⁶It should be stressed that in the (very) narrow interval $f_m < f < 1$, there is no collinear state which is locally stable.

($0 < \theta < \pi/2$) is allowed: so, it is not possible to change with continuity the value of θ which minimizes $E(\theta)$, from $\theta = 0$ (\perp state) to $\theta = \pi/2$ (\parallel state). Nevertheless, this result may be modified for two reasons: (i) the introduction of higher order anisotropies [25], and (ii) an increase of the thickness [26].

In the former case, a term like $K_4 \cos^4 \theta$ is added to $E(\theta)$, and consequently $E'(\theta) = 0$ allows solutions other than $\theta = 0, \pi/2$. This means that the minimum is expected to change continuously from $\theta = 0$ to $\theta = \pi/2$, upon varying the parameters. In the latter case ($N > 1$), a possible canting of the magnetization is determined by the break of translational invariance in the perpendicular direction, due to the surface character of the easy-axis anisotropy [26], or even by the dipolar interaction itself, whose easy-plane effect varies a little from plane to plane [10].

From an experimental point of view, case (i) is much more important: in fact, the extension of the region where canting is really observed depends on K_4/K in the first case, and on Ω/J –or K/J – (two much smaller quantities) in the second one. In other words, variations of the effective anisotropy from layer to layer change the order of the RPT (from discontinuous to continuous), but the transition from \perp to \parallel keeps very sharp.

Till now, we have analyzed the order of the RPT in the parameter space, at $T = 0$. What happens if the transition is induced by an increase in temperature? We will consider the simplest case: a single monolayer in absence of fourth-order anisotropies. In fact, this is the only case which shows a first order transition in the parameter space.

We have said that spin-wave interactions lead to a renormalization of the coupling parameters: $(J, K, \Omega) \rightarrow (J(T), K(T), \Omega(T))$. Anyway, if the *form* of the Hamiltonian keeps the same, the energy of a collinear configuration: $E(\theta, T) = -(K(T) - \Omega(T)) \cos^2 \theta$ cannot have other minima than $\theta = 0, \pi/2$, and therefore no canted configuration is possible. This is the result of several authors [24, 27, 22, 28], but it does not agree with recent calculations by Chui [29]. This author claims that a single renormalization group equation for the dipolar coupling is not sufficient: under the flow of the renormalization group, the Hamiltonian would acquire an additional term which changes the order of the transition, as the introduction of a quartic anisotropy made the transition a continuous one. Unfortunately, the lack of details in Ref. [29] prevents from drawing any conclusion.

5 Domain structures at finite temperature

We have seen how the competition between \mathcal{H}_{ex} and \mathcal{H}_{dip} gives rise –at $T = 0$ – to an ordered domain structure. This network of domain walls can be seen as a two dimensional solid, described by some positional and orientational order. This solid will be seen to support long wavelength excitations (wall meandering) as well as topological excitations (dislocations and stripe rotation domain walls).

In the Ising model, a single domain wall can be considered the interface between regions of opposite magnetisation. In two dimensions, the free energy $\sigma(T)$ of such an interface goes to zero at the critical temperature T_c of the Ising model; nevertheless, it represents a one dimensional interface, which is rough at any finite T . In the present case, “rough” means delocalized. This would imply that the solid is always in a floating phase, with the walls which are depinned from the substrate.

This is the *scenario* considered by Gehring and Keskin [30], who simply replace the domain wall energy, by its free energy. Therefore $L \approx \exp[\sigma(T)/g]$ decreases with T , till

L becomes of the same order of the domain wall size: from now on, no regular structure exists and a disordered phase sets in.

Czech and Villain [5] had pointed out that a floating phase exists only at temperatures higher than $T_R \approx g$. Indeed, neighbouring walls interact both entropically and *via* the long-range dipolar coupling. At sufficiently low T , entropic interactions can be neglected and a domain wall consists of straight pieces of wall separated by kinks. The typical distance between kinks is $\ell \approx e^{2J/T}$, where $(2J)$ is the energy of a kink. A “coarse-grained” version of the system is obtained by sampling the m -th wall at the points $y_p = p\ell'$, where $\ell' < \ell$. The resulting dynamical variables are the displacements $\{u_m^p\}$ of the sampled points, with respect to the $T = 0$ positions. The value ℓ' is chosen sufficiently small so that $|u_m^p - u_m^{p+1}| \leq 1$, but sufficiently large in order to suppose the different pieces of wall as independent. Even if kinks are absent ($|u_m^p - u_m^{p+1}| \equiv 0$), the rigid translation of a wall costs dipolar energy. The final expression for the free energy writes [5]:

$$F = \sum_{m,p} \left[J_{\parallel} (u_m^p - u_m^{p-1})^2 + J_{\perp} (u_m^p - u_{m-1}^p)^2 \right] , \quad (32)$$

which looks like an anisotropic Solid on Solid (SOS) model. The coupling constant in the wall direction ($J_{\parallel} \equiv T \ln(\ell/\ell')$) is determined by the kink energy ($= T \ln \ell = 2J$) and by the entropy $S = \ln \ell'$ of each piece of wall. On the other side, the coupling constant $J_{\perp} \equiv g\ell'/L^2$ is nothing but $(\partial^2/\partial L^2)(-g \ln L)$ times ℓ' .

It is easily seen that $J_{\parallel} \approx J_{\perp} \approx g$ when $T \approx g$: this means that the roughening temperature of the solid of domain walls is not $T_R = 0$, but rather $T_R \approx g$. At smaller temperatures, Czech and Villain suggest that L changes discontinuously (because of the discrete nature of the lattice), however keeping itself a too large value to be experimentally observable. At higher temperatures the network of domain walls represents a floating solid.

We close this part by mentioning the mean-field result [5]. In this approximation, the free energy writes:

$$F_{MF} = \sum_k (T - \tilde{J}(k)) m_k m_{-k} + \tilde{c} T \sum_i m_i^4 + \dots , \quad (33)$$

where \tilde{c} is a numerical factor and $\tilde{J}(k)$ is the Fourier transform of the exchange and dipolar coupling between spins.

The value k_m which minimizes the quadratic form is of order (g/J) , so that $L(T_C) \approx J/g$, whilst $T_C = \tilde{J}(k_m) \simeq J - O(g)$. In other words, mean-field theory predicts relatively small domains at the transition (of size of order of the dipolar length) and that T_C is only slightly modified by dipolar interactions (at least in the limit $g \ll J$).

The most detailed and complete study on the different phases of a two dimensional network of domain walls has been done by Kashuba and Pokrovsky [14] and by Abanov, Kalatsky, Pokrovsky and Saslow [31] (hereafter called K2APS theory). They consider a Heisenberg model: because of the possibility of a Reorientation Phase Transition, the phase diagram is more complex. Anyway, if we neglect the phases where the order parameter is the magnetization, and we retain only the phase diagram of the two dimensional solid of domain walls, we obtain a system which does not differ too much from the Ising case. Indeed, in the limit $L \gg w$ it is possible to replace the Heisenberg domains, separated by Bloch walls, with Ising domains, separated by empty walls. Spin-wave fluctuations are taken into account through a renormalization of the different coupling parameters. (This procedure is

expected to be correct if L is much larger than the length L_R at which renormalizations stop. Since this length is given by the relation $J/L_R^2 \approx \lambda$, the above condition equals $L \gg w$.)

Once the spins have no in-plane component, the “volume” interaction in $(1/r^3)$ between domains can be reduced –through the Gauss’s theorem– to an interaction in $(1/r)$ between domain walls. This allows to pass from spin variables (\vec{S}_i) to “displacement” variables $(u(x, y))$ of the domain walls, and the energy of the system can be written as a functional of the field $u(x, y)$.

In very general terms, we can say that the energy must contain three terms: i) A compression term, expressing the response of the system to a changement of L . This term is proportional to $(\partial_x u)^2$. ii) A bending term, expressing the energy due to a curvature $(\partial_y^2 u)$ of the domain wall. iii) A stripe orientation term, expressing the existence of preferred directions for domain walls. This term, proportional to $(\partial_y u)^2$, has an extreme importance because its presence drastically alters the behaviour of the system.

By taking into account all the above mentioned terms, we obtain [14, 31]:

$$E_S = \int \int dx dy \left[\frac{K_S}{2} \left(\frac{\partial u}{\partial x} \right)^2 + \frac{\mu}{2} \left(\frac{\partial^2 u}{\partial y^2} \right)^2 + \frac{\nu}{2} \left(\frac{\partial u}{\partial y} \right)^2 \right]. \quad (34)$$

The first term is the “continuum” version of the term proportional to J_\perp in the SOS Hamiltonian by Czech and Villain [5]. In fact, according to K2APS theory, $K_S \approx \Omega/L$, which becomes, after the transformation $x \rightarrow Lx$, $K_S \approx \Omega/L^2$. This expression equals J_\perp , once we have put $\ell' = 1$.

We could therefore be induced to conclude that the third term corresponds to the one proportional to J_\parallel in the same SOS Hamiltonian. Indeed, the question is more subtle, as suggested by the fact that ν is not simply proportional to the domain wall energy. In fact, since $\theta \equiv \partial_y u$ is the local orientation of the domain wall, in absence of anisotropic terms the energy E_S can not explicitly depends on it.

The microscopic nature of ν is the following: once we have fixed all the parameters, the domain wall energy per unit length depends on the orientation of the wall itself. If we consider an Ising model on a square lattice, a domain wall in the direction (0,1) –or (1,0)– is less expensive that the same domain wall in the (1,1) direction: $E_{dw}(0, 1) = 2J/a$ and $E_{dw}(1, 1) = 2J/(a/\sqrt{2}) = \sqrt{2}E_{dw}(0, 1)$, where we have explicitly written the lattice constant a . So, on a square lattice we will have a stripe orientation energy with periodicity $(\pi/2)$. Its expansion in the vicinity of a minimum gives rise to the third term of E_S .

The correct expression of ν for the Heisenberg model is given in [14]: $\nu = \nu_{ex} = \frac{1}{L} \left(\frac{J}{w^3} \right)$, where the factor $(1/L)$ simply translates the continuum description of walls at distance L . We want to point out that ν may have also a different origin: an in-plane anisotropy, for example. It is easily seen that in this case $\nu = \nu_{an} \approx K_\parallel w/L$, where K_\parallel is the single-ion in-plane anisotropy. So, $\nu_{an} > \nu_{ex}$ if $(K_\parallel/\lambda) > (\lambda/J)$, a condition which may be fulfilled in some systems. Furthermore, the periodicity of ν_{an} may be different from that of ν_{ex} (see the end of this section).

Finally, the bending energy term (proportional to μ) –which was neglected in [5]– is proportional to the square of the curvature $(\partial_y^2 u)$, because of the required invariance $u \rightarrow -u$.

Once we have explained the origin of the energy E_S , let us discuss the T -dependent behaviour of our system. Two quantities are of interest: the mean square fluctuations of the position $\langle u^2 \rangle$, and the mean square fluctuations of the angular deviations $\langle (\partial_y u)^2 \rangle$. The

first ones are given by:

$$\langle u^2 \rangle = T \int \int \frac{dp_x dp_y}{K_S p_x^2 + \nu p_y^2 + \mu p_y^4} = \frac{2T}{\sqrt{K_S}} \int \frac{dp_y}{\sqrt{\nu p_y^2 + \mu p_y^4}} \arctan \sqrt{\frac{K_S}{\nu p_y^2 + \mu p_y^4}}. \quad (35)$$

Since an ultraviolet cutoff is naturally supplied by the discrete nature of the system ($|p_y| < 1$), we are mainly concerned with a possible infrared divergence. When $p_y \rightarrow 0$, the integral has always a divergence: a logarithmic one if $\nu \neq 0$ and a power-like one if $\nu = 0$. The difference is relevant, because the calculation of the correlation function $\langle u(0)u(\vec{r}) \rangle$ shows a power-like decay, and therefore the existence of a quasi long-range order (QLRO), solely in the first case: in absence of a stripe orientation energy, domains are completely delocalised and the derivative $(\partial_x u)$ has even no meaning. The calculation of $\langle (\partial_y u)^2 \rangle$ simply needs to add a factor (p_y^2) at the numerator of the previous integral: because of this factor, the angular deviations are always finite, even if $\nu = 0$.

By summarizing: in the “real” case $\nu \neq 0$, there is an orientational long-range order (LRO) and a positional QLRO. A magnetic domain phase with these properties has been indeed observed by Allenspach and Bischof [21] in an ultrathin film of Fe/Cu(100), with stripe domains oriented along a high symmetry direction in the plane.

The absence of LRO for the position of domain walls apparently clashes with the results by Czech and Villain. Anyway, no real contradiction exists: at the extremely low temperatures considered in [5] ($T < \Omega$), excitations in the Ising model are given by kinks, and a discrete description of domain walls is necessary. In this case, we obtain the SOS Hamiltonian (32), to which corresponds a finite depinning temperature. Conversely, the picture given by K2APS theory is a continuous one, valid at not too low temperatures ($T > \Omega$), where even the Czech and Villain’s theory foresees a floating phase.

Let us come back to Eq. (34): at sufficiently large lengthscales, the higher order term $(\partial_y^2 u)^2$ is negligible, and E_S keeps an anisotropic quadratic form. This energy is well known [32] to correspond to an algebraical decay of the fluctuations of the order parameter. The corresponding solid is called a smectic crystal by K2APS: the QLRO is destroyed by the proliferation of pairs of dislocations,⁷ and the corresponding critical temperature writes according to the Kosterlitz-Thouless expression [33] (P stands for “positional”): $T_P = \frac{1}{8\pi} K' b^2 = \frac{\sqrt{K_S \nu}}{2\pi} L^2$, where $K' = \sqrt{K_S \nu}$ is the (renormalized) effective elastic constant and $b = 2L$ is the Burger vector corresponding to the dislocation.

Above T_P the compression coefficient vanishes and domain walls are delocalized. What happens to the orientational order parameter (hereafter explicitly written through the angle θ)? The bending energy rewrites as $\frac{\mu}{2} (\partial_{y'} \theta)^2$, whilst for the orientational stripe energy the complete expression will be used: $\frac{\nu}{16} (1 - \cos 4\theta)$. We must also add a term of the form $\frac{\kappa}{2} (\partial_{x'} \theta)^2$, which would correspond to $\frac{\kappa}{2} (\partial_{x_y}^2 u)^2$ in the “old” coordinates and which was irrelevant to determine the large scale properties of E_S . Finally, we will have [31]:

$$E_N = \int \int dx' dy' \left[\frac{\kappa}{2} \left(\frac{\partial \theta}{\partial x'} \right)^2 + \frac{\mu}{2} \left(\frac{\partial \theta}{\partial y'} \right)^2 + \frac{\nu}{16} (1 - \cos 4\theta) \right], \quad (36)$$

where (x', y') are local coordinates.

⁷A pair of dislocations of opposite Burger vectors rises from the “collapse” of a piece of two neighbouring walls.

An important point is the sign of κ , which may be positive or negative: its mean-field calculation gives $\kappa_{mf} \approx -\Omega L$, but both thermal fluctuations and corrections due to free dislocations give [31] a positive contribution to κ . An orientational ordered phase will exist at $T > T_P$, only if $\kappa > 0$. In this case, because of the anisotropic term proportional to ν —which assures an orientational LRO— the order parameter θ has a discrete Z_2 symmetry: domain walls, in a square lattice, can be directed either along a given direction, or along the direction perpendicular to it. For this reason, this phase has been called Ising nematic phase. At temperatures slightly larger than T_P , the two directions are not equally probable and the Z_2 symmetry is broken. Two contiguous regions where domains are directed along the two different “easy-axes”, are separated by a so-called stripe rotation domain wall, studied in detail by Abanov et al. [31]. At a temperature $T_0 > T_P$ we therefore expect an Ising-type transition towards a liquid phase. Since there will be two (mutually perpendicular) directions which are more likely, this phase is called a “tetragonal liquid”. In this phase, square domains should be still observable, with a magnetization inside each domain which is perpendicular to the surface. This phase may evolve towards a planar phase, *via* a Reorientation Phase Transition, or directly towards a paramagnetic phase.

For a negative κ , the Ising nematic phase does not exist and there is [31] a first-order phase transition between the (smectic) crystal and the (tetragonal) liquid.

Till now, we have supposed that the anisotropic stripe energy has a $(\pi/2)$ -periodicity and therefore domains orient preferentially along two perpendicular directions. We can suppose that in certain cases the in-plane symmetry is reduced, for example because of an uniaxial distorsion, which determines a quadratic spin anisotropy (which looks like $\mathcal{H}_{an}^{\parallel} = -\frac{K_{\parallel}}{2} \int d\vec{x} S_y^2$) or a modification of \mathcal{H}_{ex} such that the exchange coupling J differs in the two spatial directions x and y . In both cases, the existence and the nature of the smectic-nematic transition at $T = T_P$ are not changed, because for $T < T_P$ only the expansion at small θ of the stripe energy is relevant, not its global periodicity. Conversely, for $T > T_P$ we are interested in the orientational order, and for a π -periodicity the resulting scenario is completely different. In fact, in this case there is only one preferred direction for domain walls and no symmetry breaking transition may take place at $T = T_O$. The system behaves as a nematic in an electric field. The orientational order is not destroyed, till a “magnetic” transition towards a planar phase, or a paramagnetic phase, occurs.

It is interesting to compare the given picture with the results of a Monte Carlo simulation on an Ising square model, due to Booth et al. [34]. The authors study a 64×64 spins sample with a ratio $J/\Omega \simeq 9$, such that $L \simeq 8$ at $T = 0$. They single out three different phases, through the existence of two maxima in the specific heat C . By the comparison with Monte Carlo configurations at different temperatures, they conclude that there is (I) A low- T phase with an orientational order of domain walls; (II) A middle- T phase with no orientational order, but still well defined domains; (III) A high- T phase, which is fully disordered.

Authors suggest that (I) resembles the smectic crystal phase, and (II) the tetragonal liquid one. Since their model can not undergo a RPT, (III) is the paramagnetic phase. Authors also add that such parallel suffers a weak point: the transition between (I) and (II) seems to be continuous, whilst the transition between the smectic crystal and the tetragonal liquid would be first-order, or there would be a further phase (the Ising nematic one) in between.

In this respect, some comments are in order. Authors [34] study only the orientational order, and no details are given about the positional one. Besides, domain size is rather

small and dislocations are not visible in the figures reported in [34]. However, the most important point is the nature of the transition between the smectic crystal and the Ising nematic phase. This transition corresponds to the loss of the positional QLRO and has the nature of a Berezinsky-Kosterlitz-Thouless transition. Such transition, linked to the unbounding of pairs of dislocations, is not “visible” through the specific heat C , because it corresponds to an unobservable essential singularity and C has no maximum for $T = T_P$.

We argue that the observed phase transition between (I) and (II) should correspond to the transition between the Ising nematic phase and the tetragonal liquid.

6 Conclusions

In this paper we have reviewed theoretical results on magnetic domains in ultrathin films. Two-dimensional domain structures have a more fundamental meaning than the corresponding ones in bulk systems: in fact, in three dimensions domains strongly depend on the shape of the sample, and in a given sample are generally present for any direction of the magnetization⁸. Conversely, in two dimensions shape effects are negligible in the thermodynamic limit, but domains are absent if the magnetization lies in the film plane. Furthermore, the interplay between uniaxial anisotropy and shape anisotropy possibly determines a reorientation transition: domains are expected to appear just before such transition takes place (upon increasing the thickness and/or the temperature).

The main limit of the present article is to have neglected the possible application of a magnetic field. Because of the “reduced” quantity of matter, in a thin film the energy gain of a domain structure with respect to a collinear state is extremely low (see Fig. 1b). As a consequence of this, a lower magnetic field is necessary to obtain a single domain state.

A theoretical comprehension of the T -dependence of the network of domain walls has received an important impulsion with the K2APS theory, but the possibility to discover in a real system the proposed sequence of phase transitions is limited by the smallness of the region of temperatures where domains are observable. Furthermore, comparison of theoretical predictions with experimental data is made more difficult in ultrathin films by the increased importance of defects (dislocations and vacancies) and by the roughness of the interfaces.

Finally, simulation data start to be available, but they often concern Ising systems, and domain sizes are generally far from being realistic.

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⁸With the exception of a toroidal sample.

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